

FORM PTO 1390

(REV 5-98)

U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE

ATTORNEY DOCKET NUMBER

2001_0479A

**TRANSMITTAL LETTER TO THE UNITED STATES
DESIGNATED/ELECTED OFFICE (DO/EO/US)
CONCERNING A FILING UNDER 35 U.S.C. §371**

U.S. APPLICATION NO.

(If known, enter PCT No.)

NEW

09/830358

International Application No.
PCT/FR99/02036

International Filing Date
August 24, 1999

Priority Date Claimed
October 26, 1998

Title of Invention

LIQUID COMPOSITION WITH FUNGICIDAL, BACTERICIDAL OR BACTERIOSTATIC ACTIVITY

Applicant(s) For DO/EO/US

Ghislain DUFAU, Michel BARSACQ, and Gérard MOLLA

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. ☒ This is a **FIRST** submission of items concerning a filing under 35 U.S.C. §371.
2. ☐ This is a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. §371.
3. ☒ This express request to begin national examination procedures (35 U.S.C. §371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. §371(b) and PCT Articles 22 and 39(1).
4. ☒ A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date.
5. ☒ A copy of the International Application as filed (35 U.S.C. §371(c)(2))
 - a. ☒ is transmitted herewith (required only if not transmitted by the International Bureau). (See WO 00/24259)
 - b. ☐ has been transmitted by the International Bureau.
 - c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/US)
6. ☒ A translation of the International Application into English (35 U.S.C. §371(c)(2)).
7. ☒ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. §371(c)(3)).
 - a. ☐ are transmitted herewith (required only if not transmitted by the International Bureau).
 - b. ☐ have been transmitted by the International Bureau.
 - c. ☐ have not been made; however, the time limit for making such amendments has NOT expired.
 - d. ☒ have not been made and will not be made.
8. ☐ A translation of the amendments to the claims under PCT Article 19.
9. ☒ An oath or declaration of the inventor(s) (35 U.S.C. §371(c)(4)).
10. ☐ A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. §371(c)(5)).

Items 11. to 14. below concern other document(s) or information included:

11. ☒ An Information Disclosure Statement under 37 CFR 1.97 and 1.98.
12. ☒ An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.
13. ☒ A **FIRST** preliminary amendment.
☐ A **SECOND** or **SUBSEQUENT** preliminary amendment.
14. ☒ Other items or information: (a) Form PCT/RO/101; (b) Form PCT/IB/304; and (c) Form PCT/IPEA/416 with attached International Preliminary Examination Report

U.S. APPLICATION NO. 09/830358
NEW

INTERNATIONAL APPLICATION NO.
PCT/FR99/02036

ATTORNEY'S DOCKET NO.
2001 0479A

15. [X] The following fees are submitted

BASIC NATIONAL FEE (37 CFR 1.492(a)(1)-(5)):

Neither international preliminary examination fee nor international search fee paid to USPTO and International Search Report not prepared by the EPO or JPO \$1000.00
International Search Report has been prepared by the EPO or JPO \$ 860.00
International preliminary examination fee not paid to USPTO but international search paid to USPTO \$ 710.00
International preliminary examination fee paid to USPTO but claims did not satisfy provisions of PCT Article 33(1)-(4) \$ 690.00
International preliminary examination fee paid to USPTO and all claims satisfied provisions of PCT Article 33(1)-(4) \$ 100.00

ENTER APPROPRIATE BASIC FEE AMOUNT =

\$860.00

Surcharge of \$130.00 for furnishing the oath or declaration later than [] 20 [] 30 months from the earliest claimed priority date (37 CFR 1.492(e)).

\$

| Claims | Number Filed | Number Extra | Rate | |
|---|--------------|--------------|------------|----------|
| Total Claims | 19 -20 = | 0 | X \$18.00 | \$ |
| Independent Claims | 5 - 3 = | 2 | X \$80.00 | \$160.00 |
| Multiple dependent claim(s) (if applicable) | | | + \$270.00 | \$ |

TOTAL OF ABOVE CALCULATIONS =

\$1020.00

☐ Small Entity Status is hereby asserted. Above fees are reduced by 1/2.

\$

SUBTOTAL =

\$1020.00

Processing fee of \$130.00 for furnishing the English translation later than [] 20 [] 30 months from the earliest claimed priority date (37 CFR 1.492(f)).

+

\$

TOTAL NATIONAL FEE =

\$1020.00

Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). \$40 per property +

\$40.00

TOTAL FEES ENCLOSED =

\$1060.00

Amount to be refunded \$

Amount to be charged \$

a. [X] A check in the amount of \$1060.00 to cover the above fees is enclosed. A duplicate copy of this form is enclosed.

b. ☐ Please charge my Deposit Account No. 23-0975 in the amount of \$_____ to cover the above fees. A duplicate copy of this sheet is enclosed.

c. ☐ The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. 23-0975.

NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.

19. CORRESPONDENCE ADDRESS



000513

PATENT TRADEMARK OFFICE

By: *Michael R. Davis*

Michael R. Davis
Registration No. 25,134

WENDEROTH, LIND & PONACK, L.L.P.
2033 "K" Street, N.W., Suite 800
Washington, D.C. 20006-1021
Phone: (202) 721-9200
Fax: (202) 721-8250

April 25, 2001

[CHECK NO. *44155*]

[2001_0479A]

THE COMMISSIONER IS AUTHORIZED
TO CHARGE ANY DEFICIENCY IN THE
FEES FOR THIS PAPER TO DEPOSIT
ACCOUNT NO. 23-0975

09/830358

JC08 Rec'd PCT/PTO 25 APR 2001

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re application of :
Ghislain DUFAU et al. : Attn: **BOX PCT**
Serial No. NEW : **Docket No. 2001_0479A**
Filed April 25, 2001 :

LIQUID COMPOSITION WITH FUNGICIDAL,
BACTERICIDAL OR BACTERIOSTATIC
ACTIVITY
[Corresponding to PCT/FR99/02036
Filed August 24, 1999]

PRELIMINARY AMENDMENT

Assistant Commissioner for Patents,
Washington, DC 20231

Sir:

Please amend the above-identified application as follows:

IN THE SPECIFICATION

Page 1, after the title of the invention, please insert:

--This application is a 371 application of PCT/FR99/02036, filed August 24, 1999.--

IN THE CLAIMS

Cancel, without prejudice to the subject matter involved, claims 1-19.

Please add the following new claims:

--20. A fungicidal, bactericidal or bacteriostic plant-protection composition comprising at least one inorganic salt, one oxide or one hydroxide of copper in suspension in an aqueous emulsion of at least one terpenic derivative.

21. The composition of Claim 20, wherein said inorganic salt, oxide or hydroxide of copper is selected from copper hydroxide, copper oxichloride, copper carbonate, copper(I) oxide and mixtures thereof.

22. The composition of Claim 20, wherein said inorganic salt, oxide or hydroxide of copper is copper hydroxide.

23. The composition of claim 20, wherein copper is present from 200 to 600 g/l, in the form of an inorganic salt, oxide or hydroxide of copper.

24. The composition of claim 20, wherein the diameter of the particles of said inorganic salt, oxide or hydroxide of copper is not greater than 6 μm .

25. The composition of claim 20, wherein said terpenic derivative is a monoterpene or a mixture of monoterpenes.

26. The composition of claim 20, wherein said terpenic derivative is selected from terpenic hydrocarbons, oxidized derivatives of terpenic hydrocarbons, terpenic alcohols, terpenic aldehydes and ketones and mixtures thereof.

27. The composition of claim 20, wherein said terpenic derivative is a mixture of terpenic hydrocarbons and terpenic alcohols.

28. The composition of claim 20, wherein said terpenic derivative is an essential oil.

29. The composition of claim 20, wherein said terpenic derivative is a pine oil.

30. The composition of claim 20, wherein said terpenic derivative is a pine oil containing 90% of terpenic alcohols.

31. The composition of claim 20, wherein said terpenic derivative(s) is (are) present from 50 to 400 g/l.

32. The composition of claim 20, further comprising at least one surfactant.

33. The composition of claim 20, further comprising from 20 to 100 g/l of surfactant(s).

34. The composition of claim 20, further comprising a surfactant selected from:

- ethoxylated fatty acids,
- ethoxylated fatty alcohols,
- calcium alkylbenzenesulfonate,
- alkylnaphthalenesulfonates,
- ethoxylated alkylphenols,
- EO/PO block copolymers,
- PO/EO block copolymers,
- diisopropylnaphthalenesulfonates,
- dimethylnaphthalenesulfonates,
- di-n-butylnaphthalenesulfonates,
- ethoxylated dodecylphenols,
- sodium dodecylbenzenesulfonate,
- phosphoric esters of alkyl polyethers (acid forms and/or salts),
- phosphoric esters of ethoxylated arylphenols (acid forms and/or salts),
- phosphoric esters of ethoxylated polyarylphenols (acid forms and/or salts),
- ethoxylated castor oil,
- isopropylnaphthalenesulfonates,
- lignosulfonates,

- methyldinaphthalenesulfonates,
- methylnaphthalenesulfonates,
- n-butylnaphthalenesulfonates,
- ethoxylated octylphenols,
- phenyl sulfonates,
- polyalkylnaphthylmethanesulfonates,
- polyacrylates,
- ethoxylated polyarylphenols,
- polycarboxylates,
- polyvinylpyrrolidone and derivatives thereof,
- salts of sulfonated cresol-formalin condensates,
- salts of condensates of naphthalenesulfonic acid,
- salts of acrylic acid-acrylic ester copolymers,
- salts of maleic acid-olefin copolymers,
- salts of maleic anhydride-isobutylene copolymers,
- ethoxylated alkylphenol sulfates,
- ethoxylated polyarylphenol sulfates,
- sulfosuccinates,
- taurates, and
- ethoxylated tristyrylphenols.

35. A method of preparing a fungicidal, bactericidal or bacteriostic plant-protection composition comprising at least one inorganic salt, one oxide or one hydroxide of copper in suspension in an aqueous emulsion of at least one terpenic derivative, comprising the step of micronizing said inorganic salt, oxide or hydroxide of copper and other ingredients of the composition until a stable homogeneous suspension is obtained in which the size of the particles is less than 6 μm .

36. A method of preparing a fungicidal, bactericidal or bacteriostic plant-protection composition comprising at least one inorganic salt, one oxide or one hydroxide of copper in suspension in an aqueous emulsion of at least one terpenic derivative, comprising the step of admixing said inorganic salt, oxide or hydroxide of copper, having a diameter not greater than 6 μm , with other

ingredients of the composition until a stable homogeneous suspension is obtained.

37. A method of enhancing the efficacy of an inorganic salt, oxide or hydroxide of copper in a plant-protection composition, comprising combining said inorganic salt, oxide or hydroxide of copper with a terpenic derivative.

38. A method of treating plants with a product based on an inorganic salt, oxide or hydroxide of copper, comprising spraying an effective quantity of a plant-protection mixture prepared by mixing, in aqueous form, a composition of an inorganic salt, oxide or hydroxide of copper in suspension in an aqueous emulsion containing at least one terpenic derivative, on the plant to be treated.--


REMARKS

Original claims 1-19 have been canceled in favor of new claims 20-38, respectively, to avoid the multiple dependency of the original claims.

Respectfully submitted,

Ghislain DUFAU et al.

By


Michael R. Davis

Registration No. 25,134

Attorney for Applicants

MRD/aeH
Washington, D.C. 20006-1021
Telephone (202) 721-8200
Facsimile (202) 721-8250
April 25, 2001

THE COMMISSIONER IS AUTHORIZED
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FEES FOR THIS PAPER TO DEPOSIT
ACCOUNT NO. 23-0975

Liquid composition with fungicidal, bactericidal or
bacteriostatic activity

The subject of the present invention is a
5 liquid composition with fungicidal, bacteriostatic or
bactericidal activity, and methods for the preparation
and use of the composition.

It has long been known to use
phytopharmaceutical products based on inorganic salts,
10 oxides or hydroxides of copper, in particular for their
fungicidal properties (vine downy mildew and the like)
but also for their bactericidal power (bacterial canker
of peach and apricot trees, bacteriosis of apple and
pear trees caused by pseudomonas, and the like) or
15 their bacteriostatic power which prevents bacterial
diseases from becoming established.

The fungicidal and bactericidal activity of
copper depends on the nature of the copper compound and
on the quality of its manufacture.

20 The "Bordeaux mixture" has been used for over a
century for the treatment of grape vine. First prepared
by the viticulturist, and then manufactured
industrially, it is obtained by accurately neutralizing
a solution of copper sulfate with a milk of lime. This
25 mixture, brought to neutrality (pH = 7) is then dried,
ground and micronized.

Among the other products based on copper, there
may be mentioned copper oxichloride, copper hydroxide,
copper carbonate, copper(I) oxide, and the like.

30 These inorganic salts, oxides or hydroxides of
copper, used alone or in combination with other
compounds, are generally provided in the form of
wetttable powders, dispersible granules, suspension
concentrates, dustable powders, and the like.

35 The inorganic salts, oxides or hydroxides of
copper may also be combined with inorganic fungicides,
in particular sulfur, or organic fungicides to form, in
the latter case, organocopper compounds.

Among the organic active substances, there may be mentioned in particular folpel, maneb, mancozeb, propineb, zineb, cymoxanil, metiram-zinc.

5 The formulations are provided in the forms described above.

10 With the exception of the dustable powders, the other compositions generally comprise surfactants, wetting agents, dispersing agents, emulsifiers, antifoams and the like, which contribute to the stability of the formulations and then to the use of the products, and in particular to their dispersion in water for the production of mixtures for treatment.

15 In addition to the choice of inorganic salts, oxides or hydroxides of copper and to the selection of surfactants, formulators seek to enhance the efficacy of the products because the degree of protection from a copper compound against attacks by fungi and bacteria is closely related to its capacity to saturate the surface of the plant by forming a microscopic film of particles. Adherence to the plant followed by resistance to strong rain is also one of the objectives sought by the manufacturers.

20 For some time now, and in particular for the treatment of grape vine, many products using copper hydroxide as active substance have been appearing on the market.

The optimization of the formulations based on copper hydroxide, in particular of the suspension concentrates, has been carried out by:

30 - the search for a specific structure for the particles: crystallized fine needles, acicular structure;

- the improvement in the reduction of their size obtained by micronization or by a chemical method
35 for manufacture of copper hydroxide, it being possible for said size to reach from 0.3 to 0.4 μm ;

- the choice of the surfactants and co-formulants capable of stabilizing the formulation and of increasing the efficacy thereof.

FR 2 599 592 describes liquid formulations for the preventive and curative treatment of cryptogamic diseases of the oidium type comprising a lipophilic inorganic active substance consisting of fine ground or
5 micronized sulfur, in suspension in a liquid composed of a mixture of pine oil and water, the pine oil enhancing the efficacy of the sulfur.

The work by the inventors which has led to the present invention has made it possible to establish
10 that the efficacy of inorganic salts, oxides or hydroxides of copper can, surprisingly, be enhanced when these were combined with a terpenic derivative.

This discovery is unexpected insofar as persons skilled in the art did not expect the efficacy of
15 inorganic salts, oxides or hydroxides of copper to be enhanced by the addition of terpenic derivatives because of the difference in physicochemical nature between the inorganic salts, oxides or hydroxides of copper, on the one hand, and the sulfur metal species,
20 on the other hand, in particular because of the fact that sulfur is essentially lipophilic, while the inorganic salts, oxides or hydroxides of copper are essentially hydrophilic.

The subject of the invention is a fungicidal,
25 bactericidal or bacteriostatic plant-protection composition comprising at least one inorganic salt, one oxide or one hydroxide of copper in suspension in an aqueous emulsion of at least one terpenic derivative.

The aqueous emulsions also cover the
30 microemulsions.

The inorganic salt, oxide or hydroxide of copper consists of one or of a mixture of those mentioned above, copper hydroxide ($\text{Cu}(\text{OH})_2$) being preferred.

35 The terpenic derivatives for the purposes of the present invention are organic molecules containing ten carbon atoms in their structure.

They are therefore essentially monoterpenes.

The terpenic derivatives may be acyclic, monocyclic or bicyclic.

The following examples may be mentioned in particular:

- 5 1) the terpenic hydrocarbons:
 - a) acyclic terpenic hydrocarbons: myrcene, alloocimene, and the like;
 - b) monocyclic terpenic hydrocarbons: dipentene, terpinolene, p-cymene, limonene, and the like;
 - 10 c) bicyclic terpenic hydrocarbons: α -pinene, β -pinene or δ -3-carene, and the like;
- 2) the following compounds:
 - a- the oxidized derivatives: cineols;
 - b- the terpenic alcohols: borneol, fenchol,
15 menthanol, terpineols, geraniol, 1-terpinen-4-ol, and the like;
 - c- the aldehydes and ketones: camphor, fenchone;
 - 3) the mixtures of the products cited above;
 - 20 4) the essential oils containing the above mixtures in various proportions, for example *Malalencia alternifolia* essential oil (or tea-tree oil);
 - 5) the pine oils of natural or synthetic origin which are defined as being mixtures of terpenic
25 hydrocarbons and alcohols.

Pine oil containing 90% of terpenic alcohols is most particularly preferred.

- The suspension according to the invention advantageously also contains at least one surfactant
- 30 for its emulsifying, wetting, crystal growth inhibiting properties, and the like.

It is possible to use an anionic, cationic, amphoteric, zwitterionic and/or nonionic surfactant.

- The anionic and nonionic surfactants, alone or
- 35 in the form of a mixture, are preferred.

The following compounds are particularly well suited to the aim of the invention:

- ethoxylated fatty acids,
- ethoxylated fatty alcohols,

- calcium alkylbenzenesulfonate,
- alkyl-naphthalenesulfonates,
- ethoxylated alkylphenols,
- 5 - EO/PO block copolymers,
- PO/EO block copolymers,
- diisopropyl-naphthalenesulfonates,
- dimethylnaphthalenesulfonates,
- di-n-butyl-naphthalenesulfonates,
- ethoxylated dodecylphenols,
- 10 - sodium dodecylbenzenesulfonate,
- phosphoric esters of alkyl polyethers (acid forms and/or salts),
- phosphoric esters of ethoxylated arylphenols (acid forms and/or salts),
- 15 - phosphoric esters of ethoxylated polyarylphenols (acid forms and/or salts),
- ethoxylated castor oil,
- isopropyl-naphthalenesulfonates,
- lignosulfonates,
- 20 - methyl-dinaphthalenesulfonates,
- methylnaphthalenesulfonates,
- n-butyl-naphthalenesulfonates,
- ethoxylated octylphenols,
- phenyl sulfonates,
- 25 - polyalkyl-naphthylmethanesulfonates,
- polyacrylates,
- ethoxylated polyarylphenols,
- polycarboxylates,
- polyvinylpyrrolidone and derivatives thereof,
- 30 - salts of sulfonated cresol-formalin condensates,
- salts of condensates of naphthalenesulfonic acid,
- 35 - salts of acrylic acid-acrylic ester copolymers,
- salts of maleic acid-olefin copolymers,
- salts of maleic anhydride-isobutylene copolymers,

- ethoxylated alkylphenol sulfates,
- ethoxylated polyarylphenol sulfates,
- sulfosuccinates,
- taurates,
- 5 - ethoxylated tristyrylphenols...

The suspension concentrates of the present invention advantageously comprise from 200 to 600 g/l, preferably 300 to 500 g/l of copper, of the inorganic
10 salt, oxide or hydroxide of copper expressed relative to the copper element. The contents of terpenes in the formulations are between 50 and 400 g/l, preferably 80 to 200 g/l.

The contents of surfactant(s) in the
15 formulations are between 20 and 100 g/l, preferably 30 to 60 g/l.

The liquid compositions of the invention, also called suspension concentrates or "flowable concentrates" may be prepared by micronizing the active
20 substance, optionally mixed with a portion or all of the other ingredients, by passing through a specific mill (for example a ball mill of the [®]DYNO-MILL type) until a stable homogeneous suspension is obtained.

Mechanical micronization may be avoided and a
25 simple mixing may be sufficient if an active substance which is already micronized either mechanically or by the method of synthesis, is used. In either case, the micronization is performed until a diameter of the particles of inorganic salts, oxides or hydroxides of
30 copper not greater than 6 μ m is obtained.

An example of copper hydroxide which may be suitable for the preparation of a composition by simple mixing is the technical copper hydroxide manufactured by NORDEUTSCHE AFFINERIE, marketed by URANIA AGROCHEM
35 GmbH.

It is also possible to use a copper hydroxide prepared in accordance with the methods described in US 3,194,749 and US 4,944,935.

The compositions of the invention have a markedly improved fungicidal, bactericidal or bacteriostatic activity compared with the compositions of the state of the art not containing a terpenic derivative.

This gain in efficacy makes it possible to reduce the quantities of copper applied to the plants during treatments.

This dose reduction is very advantageous because it makes it possible to reduce the sometimes depressive action which copper exerts on plants and its phytotoxicity toward some plants.

The subject of the invention is also the use of a terpenic derivative for enhancing the efficacy of an inorganic salt, an oxide or a hydroxide of copper in a plant-protection, in particular fungicidal, bactericidal or bacteriostatic, composition.

The subject of the invention is, furthermore, a method of treating plants with a product based on an inorganic salt, oxide or hydroxide of copper, characterized in that an effective quantity of plant-protection mixture prepared by mixing, in aqueous form, a composition of an inorganic salt, oxide or hydroxide of copper in suspension in an aqueous emulsion containing at least one terpenic derivative, is sprayed on the plant to be treated.

Examples of compositions based on an inorganic salt, oxide or hydroxide of copper and a terpene according to the invention as well as the results obtained with these compositions on vine downy mildew (*Plasmopara viticola*) will be given below.

Examples 1 to 3 of compositions according to the invention

| | Formula A | | Formula B | | Formula C | |
|--|--|-----------|-----------|-----------|-----------|-----------|
| | Content of copper: from 300 to 310 g/l | | | | | |
| Copper hydroxide | 36.76% | 501 g/l | 36.76% | 500 g/l | 36.76% | 500 g/l |
| Pine oil (containing 90% of terpenic alcohols) | 6.60% | 90 g/l | 9.78% | 133 g/l | 12.94% | 176 g/l |
| Urea | 4.00% | 54.5 g/l | 4.00% | 54.4 g/l | 4.00% | 54.4 g/l |
| TENSIOFIX® BCZ (sulfated alcohol) | 1.00% | 13.6 g/l | 1.00% | 13.6 g/l | 1.00% | 13.6 g/l |
| TENSIOFIX® LX (lignosulfonate) | 1.00% | 13.6 g/l | 1.00% | 13.6 g/l | 1.00% | 13.6 g/l |
| TENSIOFIX® D40 (cationic/nonionic surfactant) | 1.00% | 13.6 g/l | 1.00% | 13.6 g/l | 1.00% | 13.6 g/l |
| Silicone-containing antifoam | 0.05% | 0.7 g/l | 0.05% | 0.7 g/l | 0.05% | 0.7 g/l |
| BARAGEL® 24 | 1.50% | 20.5 g/l | 1.00% | 13.6 g/l | 0.50% | 6.8 g/l |
| Water | 48.09% | 656.1 g/l | 45.41% | 617.5 g/l | 42.75% | 581.4 g/l |

Active substance: Technical copper hydroxide (content of copper: 62.05%)

Composition prepared by mixing the various ingredients and then micronizing by passing through a mill of the DYNOMILL type.

TENSIOFIX®: OMNICHEM trademark

BARAGEL®: NL-CHEMICAL trademark.

Example 4: Example of composition according to the invention

| | Formula D |
|---|------------------------------|
| | Content of copper: 396.1 g/l |
| Copper hydroxide | 43.55% |
| Pine oil (containing 90% of terpenic alcohols) | 659 g/l |
| Polyarylphenol phosphate which is ethoxylated and neutralized with triethanolamine | 130.5 g/l |
| Aqueous solution containing 35% of a sodium salt of a sulfonated cresol-formalin condensate | 25.3 g/l |
| Monooethylene glycol | 74.3 g/l |
| Heteropolysaccharide of the xanthan gum type | 5.84% |
| Silicone-containing antifoam | 0.11% |
| Water | qs |
| | qs 100 |
| | qs 100 |

Active substance: Technical copper hydroxide (content of copper = 62.4%) marketed by UFRANIA AGROCHEM GmbH

- Particle size < 13 μm : 100%
- < 6.6 μm : 92.4%
- < 4.7 μm : 81.5%
- < 3.3 μm : 64.8%
- < 2.4 μm : 47.6%

Composition prepared by simple mixing of the various ingredients.

Example 5: Example of composition according to the invention

| Formula E | |
|---|----------------------------|
| | Content of copper: 407 g/l |
| Copper hydroxide | 45.00% |
| Pine oil (containing 90% of terpenic alcohols) | 691.2 g/l |
| Polyarylphenol phosphate which is ethoxylated and neutralized with triethanolamine | 7.80% |
| Aqueous solution containing 35% of a sodium salt of a sulfonated cresol-formalin condensate | 2.20% |
| Glycerol | 33.8 g/l |
| Urea | 76.8 g/l |
| Silicone-containing antifoam | 1.40% |
| Heteropolysaccharide of the xanthan gum type | 6.00% |
| Water | 21.5 g/l |
| | 92.2 g/l |
| | qs |
| | 0.125% |
| | 1.9 g/l |
| | qs 100 |
| | qs 100 |

Active substance: Technical copper hydroxide (content of copper = 62.88%)

Composition prepared by mixing the various ingredients and then micronizing by passing through a ball mill of the DYNOMILL type.

Example 6: Results of experimentation on vine downy mildew (*Plasmopara viticola*)**-1st trial: Scores on leaves**

| | Dose/ha of Product | Doses/ha | | % of damage on leaves | |
|--------------------------------------|--------------------------|----------|----------|-----------------------|-----------|
| | | Copper | Pine oil | 1st score | 2nd score |
| NTC | / | / | / | 68.75% | 97.50% |
| Formula B | 5 l | 1500 g | 665 g | 43.75% | 83.75% |
| Formula C | 5 l | 1500 g | 880 g | 40.63% | 84.38% |
| Formula A | 7.5 l | 2250 g | 675 g | 43.13% | 85.63% |
| Formula B | 7.5 l | 2250 g | 998 g | 29.38% | 70.63% |
| Formula C | 7.5 l | 2250 g | 1320 g | 26.25% | 68.13% |
| SC formulation (state of the art) | 8.8 l | 3000 g | / | 38.13% | 82.50% |
| WP formulation (state of the art) | 6 kg | 3000 g | / | 42.50% | 78.13% |

Study on young plants

Grape vine, Cabernet-Sauvignon cultivar

Trials under misting with artificial contaminations

NTC: Non treated control SC: Suspension concentrate

WP: Wettable powder

-2nd trial: Scores on bunches of grapes

| | Dose/ha of Product | Doses/ha | | 1st score | | 2nd score | |
|--------------------------------------|--------------------------|----------|-------------|-----------|-----------|-----------|-----------|
| | | Copper | Pine oil | Intensity | Frequency | Intensity | Frequency |
| NTC | / | / | / | 72.85% | 14.29% | 98.20% | 58.56% |
| Formula D | 5 l | 2050 g | 650 g | 6.41% | 0.33% | 27.50% | 2.29% |
| SC formulation (state of the art) | 6.7 l | 2030 g | / | 9.09% | 0.75% | 36.03% | 2.82% |

Study on fruit-bearing plants

Grape vine, Cabernet-Sauvignon cultivar

Trials under misting with artificial contaminations

NTC: Non treated control SC: Suspension concentrate

-3rd trial: Scores on leaves

| | Dose/ha of Product | Doses/ha | | % of damage on leaves | % of defoliating |
|--------------------------------------|--------------------------|----------|-------------|-----------------------------|---------------------|
| | | Copper | Pine oil | | |
| Formula D | 5 l | 2050 g | 650 g | 22.50% | 48.75% |
| SC formulation (state of the art) | 6.7 l | 2030 g | / | 36.25% | 60.00% |

Study on fruit-bearing plants

Grape vine, Cabernet-Sauvignon cultivar

Trials under misting with artificial contaminations

SC: Suspension concentrate

-4th trial: Scores on leaves

| | Dose/ha of Product | Doses/ha | | 1st score | | 2nd score | | 3rd score | |
|--------------------------------------|--------------------------|----------|-------------|-----------|-----------|-----------|-----------|-----------|-----------|
| | | Copper | Pine oil | Intensity | Frequency | Intensity | Frequency | Intensity | Frequency |
| NTC | / | / | / | 15.0% | 64.5% | 62.83% | 99.0% | 58.65% | 84.0% |
| Formula E | 4 l | 1630 g | 480 g | 1.8% | 6.8% | 1.63% | 16.0% | 1.47% | 14.3% |
| Formula E | 5 l | 2040 g | 600 g | 1.5% | 6.8% | 0.90% | 9.0% | 1.33% | 15.0% |
| Formula E | 6.25 l | 2540 g | 750 g | 2.2% | 8.5% | 1.22% | 11.5% | 1.16% | 11.0% |
| SC formulation (state of the art) | 7 l | 2520 g | / | 2.0% | 8.8% | 2.20% | 19.0% | 1.91% | 15.3% |

Study on fruit-bearing plants

Grape vine, Grenache

Trials under misting with artificial contaminations

NTC: Non treated control SC: Suspension concentrate

-5th trial: Scores on bunches of grapes

| | Dose/ha of Product | Doses/ha | | 1st score | | 2nd score | |
|--------------------------------------|--------------------------|----------|-------------|-----------|-----------|-----------|-----------|
| | | Copper | Pine oil | Intensity | Frequency | Intensity | Frequency |
| NTC | / | / | / | 9.90% | 57.5% | 67.70% | 99.8% |
| Formula E | 4 l | 1630 g | 480 g | 0.20% | 2.3% | 7.05% | 42.8% |
| Formula E | 5 l | 2040 g | 600 g | 0.15% | 2.6% | 7.30% | 36.3% |
| Formula E | 6.25 l | 2540 g | 750 g | 0.01% | 0.5% | 1.70% | 14.8% |
| SC formulation (state of the art) | 7 l | 2520 g | / | 9.09% | 1.5% | 4.10% | 29.5% |

Study on fruit-bearing plants

Grape vine, Grenache

Trials under misting with artificial contaminations

NTC: Non treated control SC: Suspension concentrate

-6th trial: Scores on leaves

| | Dose/ha of Product | Doses/ha | | 1st score | | 2nd score | | 3rd score | |
|---|--------------------------|----------|-------------|-----------|-----------|-----------|-----------|-----------|-----------|
| | | Copper | Pine oil | Intensity | Frequency | Intensity | Frequency | Intensity | Frequency |
| NTC | / | / | / | 24.3% | 84.5% | 27.83% | 97.8% | 27.34% | 91.8% |
| Formula E | 4 l | 1630 g | 480 g | 0.6% | 3.5% | 0.90% | 4.5% | 0.35% | 2.3% |
| Formula E | 5 l | 2040 g | 600 g | 0.2% | 2.3% | 0.23% | 2.3% | 0.17% | 1.3% |
| SC formulation (state of the art) | 7 l | 2520 g | / | 0.2% | 2.0% | 0.45% | 2.0% | 0.18% | 2.0% |

Study on fruit-bearing plants

Grape vine, Grenache

Trials under misting with artificial contaminations

NTC: Non treated control SC: Suspension concentrate

-7th trial: Scores on bunches of grapes

| | Dose/ha of Product | Doses/ha | | Intensity | Frequency |
|--------------------------------------|--------------------------|----------|-------------|-----------|-----------|
| | | Copper | Pine oil | | |
| NTC | / | / | / | 6.5% | 39.8% |
| Formula E | 4 l | 1630 g | 480 g | 0.1% | 0.8% |
| Formula E | 5 l | 2040 g | 600 g | 0.1% | 0.4% |
| Formula E | 6.25 l | 2540 g | 750 g | 0.0% | 0.4% |
| SC formulation (state of the art) | 7 l | 2520 g | / | 0.1% | 0.9% |

Study on fruit-bearing plants

Grape vine, Grenache

Trials under misting with artificial contaminations

NTC: Non treated control SC: Suspension concentrate

CLAIMS

1. Fungicidal, bactericidal or bacteriostic plant-protection composition comprising at least one
5 inorganic salt, one oxide or one hydroxide of copper in suspension in an aqueous emulsion of at least one terpenic derivative.
2. Composition according to Claim 1, characterized in that the inorganic salt, oxide or hydroxide of
10 copper is selected from copper hydroxide, copper oxichloride, copper carbonate, copper(I) oxide and mixtures thereof.
3. Composition according to Claim 1 or Claim 2, characterized in that the inorganic salt, oxide or
15 hydroxide of copper is copper hydroxide.
4. Composition according to any one of the preceding claims, characterized in that it contains from 200 to 600 g/l, advantageously from 300 to 500 g/l of copper, in the form of an inorganic salt, oxide or
20 hydroxide of copper.
5. Composition according to any one of the preceding claims, characterized in that the diameter of the particles of inorganic salts, oxides or hydroxides of copper is not greater than 6 μm .
- 25 6. Composition according to any one of the preceding claims, characterized in that the terpenic derivative is a monoterpene or a mixture of monoterpenes.
7. Composition according to any one of the
30 preceding claims, characterized in that the terpenic derivative is selected from terpenic hydrocarbons, oxidized derivatives of terpenic hydrocarbons, terpenic alcohols, terpenic aldehydes and ketones and mixtures thereof.
- 35 8. Composition according to any one of the preceding claims, characterized in that the terpenic derivative is a mixture of terpenic hydrocarbons and terpenic alcohols.

9. Composition according to any one of the preceding claims, characterized in that the terpenic derivative is an essential oil.

5 10. Composition according to any one of the preceding claims, characterized in that the terpenic derivative is a pine oil.

10 11. Composition according to any one of the preceding claims, characterized in that the terpenic derivative is a pine oil containing 90% of terpenic alcohols.

12. Composition according to any one of the preceding claims, characterized in that it contains from 50 to 400 g/l, advantageously from 80 to 200 g/l of terpenic derivative(s).

15 13. Composition according to any one of the preceding claims, characterized in that it comprises at least one surfactant.

14. Composition according to any one of the preceding claims, characterized in that it comprises 20 20 to 100 g/l, advantageously from 20 to 100 g/l of surfactant(s).

15. Composition according to any one of the preceding claims, characterized in that the surfactant is selected from:

- 25 - ethoxylated fatty acids,
- ethoxylated fatty alcohols,
- calcium alkylbenzenesulfonate,
- alkyl-naphthalenesulfonates,
- ethoxylated alkylphenols,
- 30 - EO/PO block copolymers,
- PO/EO block copolymers,
- diisopropyl-naphthalenesulfonates,
- dimethyl-naphthalenesulfonates,
- di-n-butyl-naphthalenesulfonates,
- 35 - ethoxylated dodecylphenols,
- sodium dodecylbenzenesulfonate,
- phosphoric esters of alkyl polyethers (acid forms and/or salts),

- phosphoric esters of ethoxylated arylphenols (acid forms and/or salts),
- phosphoric esters of ethoxylated polyarylphenols (acid forms and/or salts),
- 5 - ethoxylated castor oil,
- isopropyl naphthalenesulfonates,
- lignosulfonates,
- methyl dinaphthalenesulfonates,
- methyl naphthalenesulfonates,
- 10 - n-butyl naphthalenesulfonates,
- ethoxylated octylphenols,
- phenyl sulfonates,
- polyalkyl naphthylmethanesulfonates,
- polyacrylates,
- 15 - ethoxylated polyarylphenols,
- polycarboxylates,
- polyvinylpyrrolidone and derivatives thereof,
- salts of sulfonated cresol-formalin condensates,
- 20 - salts of condensates of naphthalenesulfonic acid,
- salts of acrylic acid-acrylic ester copolymers,
- 25 - salts of maleic acid-olefin copolymers,
- salts of maleic anhydride-isobutylene copolymers,
- ethoxylated alkylphenol sulfates,
- ethoxylated polyarylphenol sulfates,
- 30 - sulfosuccinates,
- taurates,
- ethoxylated tristyrilphenols...

16. Method of preparing a composition as defined in any one of the preceding claims, by the following step:

- 35 - the inorganic salt, oxide or hydroxide of copper and the other ingredients of the composition are micronized until a stable homogeneous suspension is obtained in which the size of the particles is less than 6 μm .

17. Method of preparing a composition as defined in Claims 1 to 15, characterized by the following step:

5 - an inorganic salt, an oxide or a hydroxide of copper, whose diameter is not greater than 6 μ m, is mixed with the other ingredients of the composition until a stable homogeneous suspension is obtained.

18. Use of a terpenic derivative for enhancing the efficacy of an inorganic salt, an oxide or a hydroxide of copper in a plant-protection, in particular
10 fungicidal, bactericidal or bacteriostatic, composition.

19. Method of treating plants with a product based on an inorganic salt, oxide or hydroxide of copper, characterized in that an effective quantity of plant-
15 protection mixture prepared by mixing, in aqueous form, a composition of an inorganic salt, oxide or hydroxide of copper in suspension in an aqueous emulsion containing at least one terpenic derivative is sprayed on the plant to be treated.

DECLARATION AND POWER OF ATTORNEY FOR U. S. PATENT APPLICATION

() Original () Supplemental () Substitute () PCT () Design

As a below named inventor, I hereby declare that: my residence, post office address and citizenship are as stated below next to my name; that I verily believe that I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural inventors are named below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

Title: **" Liquid composition with fungicidal, bactericidal or bacteriostatic activity "**.

of which is described and claimed in:

☒ the attached specification, or

() the specification in the application Serial No. _____, filed _____;

and with amendments through _____ (if applicable), or

☒ the specification in International Application No. PCT/**FR99/02036**, filed **24/08/99**, and as amended on _____ (if applicable).

I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment(s) referred to above.

I acknowledge my duty to disclose to the Patent and Trademark Office all information known to me to be material to patentability as defined in Title 37, Code of Federal Regulations, §1.56.

I hereby claim priority benefits under Title 35, United States Code, §119 (and §172 if this application is for a Design) of any application(s) for patent or inventor's certificate listed below and have also identified below any application for patent or inventor's certificate having a filing date before that of the application on which priority is claimed:


| COUNTRY | APPLICATION NO. | DATE OF FILING | PRIORITY CLAIMED |
|---------------|-----------------|-----------------|------------------|
| FRANCE | 98 13381 | 26/10/98 | YES |
| | | | |
| | | | |
| | | | |
| | | | |

I hereby claim the benefit under Title 35, United States Code §120 of any United States application(s) listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States application in the manner provided by the first paragraph of Title 35, United States Code §112, I acknowledge the duty to disclose information material to patentability as defined in Title 37, Code of Federal Regulations, §1.56 which occurred between the filing date of the prior application and the national or PCT international filing date of this application:

| APPLICATION SERIAL NO. | U.S. FILING DATE | STATUS: PATENTED, PENDING, ABANDONED |
|------------------------|------------------|--------------------------------------|
| | | |
| | | |
| | | |

And I hereby appoint Michael R. Davis, Reg. No. 25,134; Matthew M. Jacob, Reg. No. 25,154; Warren M. Cheek, Jr., Reg. No. 33,367; Nils Pedersen, Reg. No. 33,145; Charles R. Watts, Reg. No. 33,142; and Michael S. Huppert, Reg. No. 40,268, who together constitute the firm of WENDEROTH, LIND & PONACK, L.L.P., as well as any other attorneys and agents associated with Customer No. 000513, to prosecute this application and to transact all business in the U.S. Patent and Trademark Office connected therewith.

I hereby authorize the U.S. attorneys and agents named herein to accept and follow instructions from _____ as to any action to be taken in the U.S. Patent and Trademark Office regarding this application without direct communication between the U.S. attorneys and myself. In the event of a change in the persons from whom instructions may be taken, the U.S. attorneys named herein will be so notified by me.

| | |
|---|---|
| Direct Correspondence to Customer No.:  000513 PATENT TRADEMARK OFFICE | Direct Telephone Calls to: WENDEROTH, LIND & PONACK, L.L.P. 2033 "K" Street, N.W., Suite 800 Washington, D.C. 20006 Phone (202) 721-8200 Fax: (202) 721-8250 |
|---|---|


| | | | |
|------------------------------|---|-------------------------------------|--|
| Full Name of First Inventor | FAMILY NAME DUFAU | FIRST GIVEN NAME Chislain | SECOND GIVEN NAME |
| Residence & Citizenship | CITY DAX | STATE OR COUNTRY FRANCE | COUNTRY OF CITIZENSHIP FRANCE |
| Post Office Address | ADDRESS La Grange aux palombes - Route des artificiers - DAX - FRANCE - 40100 | CITY | STATE OR COUNTRY ZIP CODE |
| Full Name of Second Inventor | FAMILY NAME BARSACQ | FIRST GIVEN NAME Michel | SECOND GIVEN NAME |
| Residence & Citizenship | CITY DAX | STATE OR COUNTRY FRANCE | COUNTRY OF CITIZENSHIP FRANCE |
| Post Office Address | ADDRESS 6, rue de la Tannerie | CITY DAX | STATE OR COUNTRY ZIP CODE FRANCE 40100 |
| Full Name of Third Inventor | FAMILY NAME MOLLA | FIRST GIVEN NAME Gérard | SECOND GIVEN NAME |
| Residence & Citizenship | CITY DAX | STATE OR COUNTRY FRANCE | COUNTRY OF CITIZENSHIP FRANCE |
| Post Office Address | ADDRESS 3, rue Joseph Barsacq Mongis - DAX | CITY | STATE OR COUNTRY ZIP CODE FRANCE 40100 |
| Full Name of Fourth Inventor | FAMILY NAME | FIRST GIVEN NAME | SECOND GIVEN NAME |
| Residence & Citizenship | CITY | STATE OR COUNTRY | COUNTRY OF CITIZENSHIP |
| Post Office Address | ADDRESS | CITY | STATE OR COUNTRY ZIP CODE |


| | | | |
|-----------------------------|-------------|------------------|---------------------------|
| Full Name of Fifth Inventor | FAMILY NAME | FIRST GIVEN NAME | SECOND GIVEN NAME |
| Residence & Citizenship | CITY | STATE OR COUNTRY | COUNTRY OF CITIZENSHIP |
| Post Office Address | ADDRESS | CITY | STATE OR COUNTRY ZIP CODE |


| | | | |
|-----------------------------|-------------|------------------|---------------------------|
| Full Name of Sixth Inventor | FAMILY NAME | FIRST GIVEN NAME | SECOND GIVEN NAME |
| Residence & Citizenship | CITY | STATE OR COUNTRY | COUNTRY OF CITIZENSHIP |
| Post Office Address | ADDRESS | CITY | STATE OR COUNTRY ZIP CODE |

| | | | |
|-------------------------------|-------------|------------------|---------------------------|
| Full Name of Seventh Inventor | FAMILY NAME | FIRST GIVEN NAME | SECOND GIVEN NAME |
| Residence & Citizenship | CITY | STATE OR COUNTRY | COUNTRY OF CITIZENSHIP |
| Post Office Address | ADDRESS | CITY | STATE OR COUNTRY ZIP CODE |

I further declare that all statements made herein of my own knowledge are true, and that all statements on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

1st Inventor Ghislain DUFAU  Date 06/04/01

2nd Inventor Michel BARSACQ  Date 06/04/01

3rd Inventor Gérard MOLA  Date 06/04/01

4th Inventor _____ Date _____

5th Inventor _____ Date _____

6th Inventor _____ Date _____

7th Inventor _____ Date _____

The above application may be more particularly identified as follows:

U.S. Application Serial No. _____ Filing Date _____

Applicant Reference Number _____ Atty Docket No. _____

Title of Invention _____